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Microscale Power Generation based on Radiolytic Processes

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Abstract

Oregon State University, Battelle Pacific Northwest Division, and Case Western Reserve University have been investigating nuclear-to-electric power generation utilizing a technique based on closed cycle fuel cell operation. The approach being developed is to first use the decay energy of a radioisotope to generate H_2 and O_2 from water, then to utilize these species in a fuel cell to generate electricity. This method can regenerate its own reactants and operate continuously as a closed system for as long as the primary source of power, namely the radioisotope, is active. With micro engineering and fabrication techniques available today, a miniaturized integrated package of 1 cm^3 in size and producing power in the 10 mW range appears feasible in a mature design. For this project, a unique fuel cell capable of utilizing mixed reactants at room temperature has been developed. Efficiency of this early fuel cell design falls in the range between 15 and 20%. Measured power output from a test cell approached 0.45 mW for several hours with a radiation leakage rate estimated at 490 mrem/yr.

Keywords: Radioisotope Power Generation, Single Chamber Fuel Cell, Radiolysis

1 - INTRODUCTION

Common batteries have difficulty meeting the demands of moderate-to-low power applications over extended periods of time. This shortcoming results from the relatively low energy densities found in electrochemical cells. Decreasing the size of the battery exacerbates the problem since packaging becomes a larger *percent* of the total volume compared to that of larger conventional cells [1]. One potential route to solving this problem is to use a power source having a higher specific energy density. Radioisotopes represent a self-contained energy source capable of delivering modest amounts of power continuously for months-to-years (without recharging) and could possibly fulfill the requirements for a long lasting power supply. Figure 1 shows the difference in power densities of radioisotopes when compared to common battery types and combustible fuels [2]. The comparison may not be completely valid since shielding, containment, and other packaging considerations have not been included in the graph (the source material itself may only represent 1 to 10% of the power cell mass). However, a conversion efficiency to electrical power of 10% was used for both the combustible fuels and radioisotopes. Even with these considerations, the potential of nuclear decay power over conventional sources is evident. Practical miniature radioisotope power cells would have important applications including: 1.) powering MEMS, 2.) trickle charging batteries and super capacitors, and 3.) running small portable electronic devices and remotely located unattended sensors.

2 - UTILIZING RADIOISOTOPE POWER

The traditional approaches to converting radiodecay energy into electricity can be placed into one of three categories: (1) direct charging devices, (2) secondary particle generation schemes, and (3) thermal means. The first of these relies on the decay products traversing a vacuum potential of several thousand volts in order to capture significant quantities of the particle's emitted energy [3]. Surface emission is almost exclusively relied upon, thus only very thin layers of the radioactive source can be utilized [4]. Furthermore, secondary particle emission from the emitting surface leads to a loss of energy. Although innovative concepts may be able to overcome some of these difficulties, for a small-scale power module with a volume of the order of 1 cm^3, the challenges are formidable. Besides the direct charging concept, traditional conversion methods utilizing secondary particle generation and thermal approaches can be candidates for miniature power generation devices.

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An example of secondary particle generation is beta voltaic energy conversion [5]. It relies on the production of electrical power by coupling beta emissions from a thin sheet source into a semiconductor device. Direct electron-hole pair creation in the semiconductor and the use of a PN junction for charge separation generates the electrical power. Although these devices can be miniaturized, current technology devices (in the open literature) are of the order of 1 cubic inch and only produce power in the 10 – 100 microwatt range. Another secondary particle approach is radio luminescence power cells which consist of a beta source coupled to a material capable of generating photons when excited by high energy electrons [6]. The photons then go on to produce electricity in a photovoltaic cell. This two-step process may appear to be less efficient than the one-stage conversion of beta voltaics, but advantages are present due to the flexibility of arranging the source and luminescence material, and the lower radiation damage to sensitive components of the converter. The radioisotopes most frequently used in such power devices are ^{3}H and [147]Pm. Most of the recent power cells based on radio luminescence have outputs in the 10 microwatt range with efficiencies rarely exceeding 0.1%. Future designs may be able to exceed these values by making better use of the source energy and moving more toward a bulk emission rather than a surface emission process.
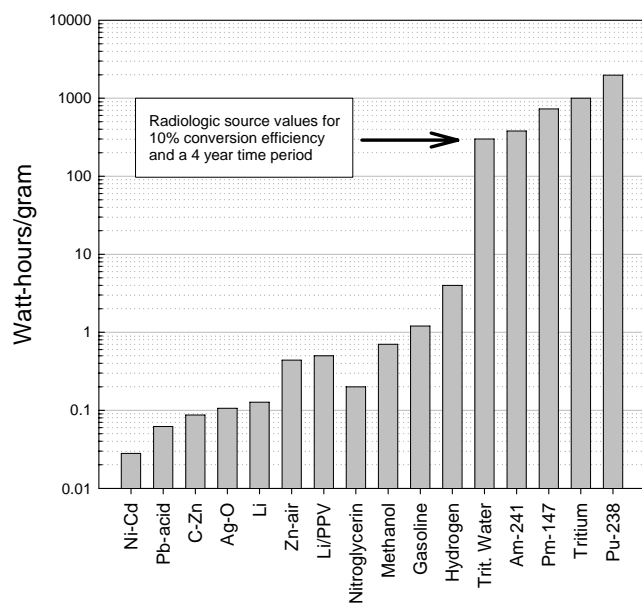

Figure 1: Comparison of various electrochemical, chemical, and radiological power sources. The latter two sources have an assumed 10% conversion efficiency to electricity (adapted from *Polymers, Phosphors, and Voltaics for Radioisotope Microbatteries).*

Generating heat through radioactive decay has been utilized for decades as small scale power sources [7]. A current application is for powering space probes to the outer planets where radiant energy from the sun is too low to be useful. NASA has invested considerable resources in developing

these power modules based on thermoelectric conversion (TEC) and current technology allows approximately 3 – 5% overall conversion efficiency with source temperatures exceeding 500 °C.

Heat production from a radioactive source is inherently volumetric, so the limitations that apply to surface emitting schemes do not apply. This allows higher overall power densities. With the miniaturization achievable using TEC technology, this method should be attractive for small-scale applications. The drawback with this method, however, is both critical and fundamental to miniaturizing the conversion process. Thermoelectric conversion requires elevated source temperatures: the higher the source temperature, the better the conversion efficiency. In order to achieve power outputs of several milliwatts in a size of one cubic centimeter, a source with a temperature in the neighborhood of 500 °C must be packaged, *with high performance insulation*, in this volume. Without thermal insulation, the heat from a small radioactive source would rapidly leak away to the surroundings resulting in the hot end of the TEC not achieving the required temperatures for efficient conversion. Due to fundamental thermal loss mechanisms, breakthroughs in thermal insulation design are needed if small package sizes are to be realized.

3 - RADIOLYTIC FUEL CELL APPROACH

During the late 1950's and early 1960's, radiochemically regenerative fuel cells were investigated for the purpose of generating large-scale power [8]. In principle, the central concept of this prior work was to use high energy decay products to convert a stable species into fuel cell reactants. The fuel cell, operating normally with separated anode and cathode chambers, would then generate electricity and produce the stable product species thus completing a regenerative cycle. The basic concept was explored for large-scale power production (in the 1 to 10 kW range). The essential difference between the electrochemical method of nuclear power production and conventional approaches is that in the former case, a chemical intermediary is involved thus isolating the power producing elements from the radioactive source. Several chemical systems were studied for this concept including the water cycle, the ferric ion cycle, and the ozone cycle [9]. This latter case was considered for space power systems at the 100 to 1000 Watt-electric range.

4 - CONFIGURATION AND EXPERIMENTS

The approach reported on here uses a single chamber fuel cell [10, 11] to produce electrical power from hydrogen and oxygen generated from radiolysis. Although a number of different radioisotopes could be considered for this application, [238]Pu was the source material for the early phase of the development program due to its availability. Our specific implementation of the concept uses alpha particles from this source to split water. The original concept was envisioned as either an alpha or beta source arranged in a meso porous thick-film matrix material where water would permeate the film. Decay products within the meso porous film would dissociate a small percentage of water into

hydrogen and oxygen which would diffuse to the fuel cell to react forming water and electricity. However, the most straightforward source configuration for early development work was a particulate source immersed in water.

4.1 - Fuel Cell Development

The fuel cell ultimately used in this work went through a number of development iterations. Preliminary work was accomplished with a cell composed of a porous silver membrane (60 microns thick, 2.5 cm in diameter) acting as a selective cathode under basic conditions. The electrolyte was 0.5 M KOH. A nylon mesh was positioned immediately in back of the silver cathode followed by a platinum/Pt-black anode mesh. The platinum acted as the anode. In this type of configuration, the idea is to have the oxygen react on the selective silver cathode with hydrogen diffusing through the cathode and the nylon mesh spacer to react on the anode. This entire assembly was "clamped" together against an impermeable back surface using a magnet acting on a nickel mesh on the outside surface of the silver membrane. A wick was attached to this nickel mesh to ensure the fuel cell was in contact with the electrolyte reservoir in the bottom of the cell. An early version of this configuration is shown in the next section (Fig. 3).

Although this cell configuration generated significant power in electrolysis experiments, testing alternative designs that exposed the backside of the cell to the gas phase environment proved to be a better configuration for further study. Many variations of this exposed backside cell were tested in the laboratory for power output and consistency. Ultimately, the configuration that proved best involved a sandwich arrangement of Ag/nylon/Pt/nylon/Ag. In order for this configuration to work, the magnet clamping was also modified such that a thin gap existed between the backside cathode and the support structure. As a final modification to this, the backside silver membrane was perforated by an array of holes to increased hydrogen diffusion to the anode.

Electrolysis experiments were conducted for "cold" testing. The configuration shown in Fig. 3 had the inner cup removed and replaced with two platinum wire electrodes. An electrolyte solution of 0.5 M KOH covered the wires. Power supplied to the electrolysis electrodes was monitored for both current and voltage. With the fuel cell suspended in the gas space above the liquid, a cold test of the power generating capability of the cell was achieved. The procedure for testing involved a purge of the interior gas space with pure hydrogen followed by stopping the flow and isolating cell. The electrolysis was then initiated at 0.5, 1.0, 2.0, 3.0, 4.0 mamps in stepped increases. Fuel cell output was monitored for both current and voltage. Results of one of these experiments are shown in Fig. 2 for a load resistance of 400 ohms. What is evident in the graph is the response of the fuel cell to increasing levels of H_2 and O_2 generation. The first three increases are consistent with increasing the reactants for the cell. However, the last increase, from 3.0 mamps to 4.0 mamps indicates a strong amount of saturation existing in the

fuel cell. That is, the cell is incapable of utilizing the reactants being generated at the higher rates of evolution from the electrolysis experiment. Our hypothesis is that H_2 transport to the anode is diffusionally limited and that higher pressures would lead to increased output.
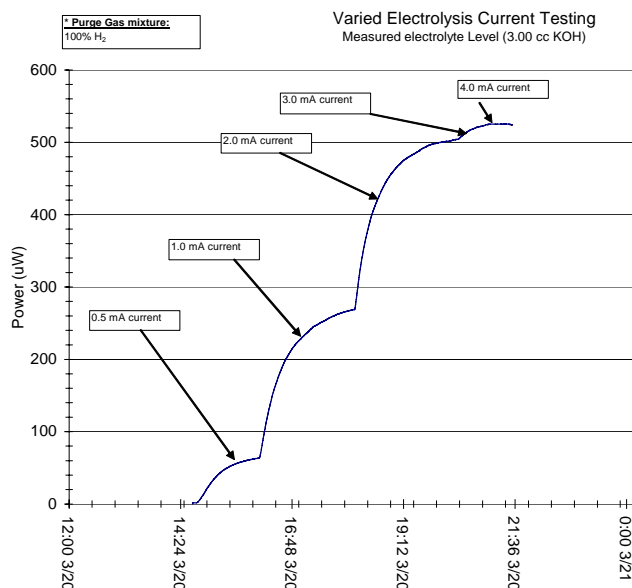

Figure 2: Fuel cell response to various levels of electrolysis current.

4.2 - Power Cell Development

An early version of the power cell configuration is pictured in Fig. 3. Although not a mature device in the sense of utilizing the envisioned source configuration, it does capture all the features of a closed cycle fuel cell operating on reactants that are continuously generated by the source. The radioisotope source material (PuO_2) is contained in an inner glass cup as a slurry of water and the particulate form of the source oxide material. The reactants generated in the cup diffuse up through the aqueous environment and enter the gas phase. A single chamber fuel cell is positioned in the gas phase right above the cup to intercept the reactants for power generation. In this design, a wick is in contact with an electrolyte reservoir in the annular region around the cup in the bottom.

The source was 1 g of PuO_2 flooded with water with a measured activity of 9 Ci/g (pure $^{238}PuO_2$ has an activity of 17 Ci/g). The dose rate from the source was estimated at 490 mrem/yr. The procedure for a power test involved loading the particulate PuO_2 into the inner cup and wetting it with distilled water. It was then placed in a larger glass beaker of 30 mm O.D. (as shown in Fig. 3) which contained the electrolyte solution in the bottom. The top seal attachment with fuel cell was lowered into the beaker until the O-ring closures where tight. Pure H_2 purge gas was flowed through the test cell at a sufficient rate to remove air and absorbed gases prior to the test. During purge, electrical attachments

were made to the fuel cell via feed-throughs in the top seal attachment (not shown in the figure). Power was monitored through the purge cycle and into the power test phase. At the beginning of the test, the purge line was clipped off and power output from the fuel cell was monitored. The hydrogen background gas has been discovered to enhance cell performance and lead to stable operation. This allows hydrogen to be present directly adjacent to the anode for cell operation, but any hydrogen consumed at the anode is replenished by the radiolytic source. This keeps the hydrogen concentration constant during cell operation.
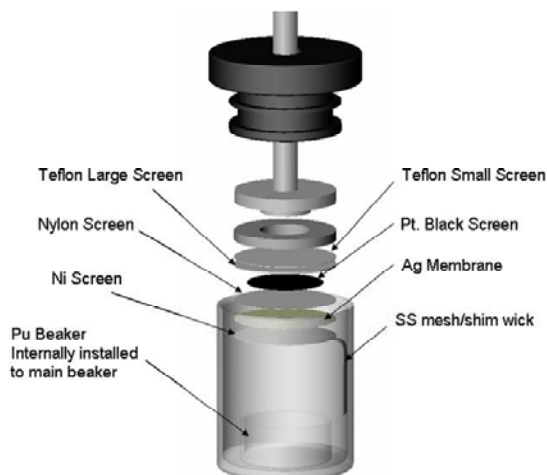

Figure 3: The test cell is comprised of an inner cup containing the PuO_2 source with a gas phase fuel cell positioned over the source.

Figure 4 presents power measurements for one of several tests conducted on the completed cell. The test cell operated near ambient temperature (only minor self-heating is present) while generating electrical power in the 0.45 mW power range. A trail-off in power was observed during longer power tests. We do not attribute this power decrease to cell damage, but rather to hydrogen leakage (through diffusion) from the cell and thus causing a loss of the ideal hydrogen-rich stoichiometry that allows the power output to be maximized. Figure 4 shows the results of one of the short term tests where power output increased from initial cell purge conditions and stabilized approximately 100 minutes.

5 - CONCLUDING REMARKS

Radioisotope power generation may find applications in areas where long term, unattended operation is a requirement at moderate-to-low power levels. One type of conversion approach, as described here, involves radiolysis coupled with a single chamber fuel cell for power production. Although the initial concept is in the early stages of development, proof-of-principle operational tests have confirmed the basic concept. Miniaturization, long term stability, lowering dose rates, and raising power levels remain as challenges in the further development of the concept.
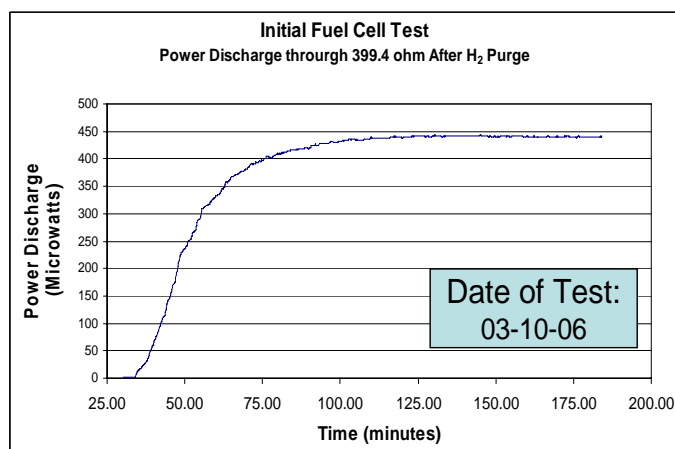

Figure 4: Power cell test results using a PuO_2 source flooded with water.

REFERENCES

[1] Handbook of Batteries, 3rd edition, Edited by David Linden and Thomas B. Reddy. McGraw-Hill , 2002, New York. Chapter 3 (written by David Linden) pg 3.21.

[2] Bower, K.E., Rutkiewic, A.F., Bower, C.C., and Yousaf, S.M., Radioisotope Microbattery Commercialization. In *Polymers, Phosphors, and Voltaics for Radioisotope Microbatteries* (Eds. K.E. Bower, et al.), 2002, p. 444, (CRC Press, New York).

[3] Miley, G.H., *Direct Conversion of Nuclear Radiation Energy*, Monograph Series on Nuclear Science and Technology, American Nuclear Society, pp. 1 – 89, 1970.

[4] Kavetsky, A.G., et al., Radioactive Materials, Ionizing Radiation Sources, and Radioluminescent Light Sources. In *Polymers, Phosphors, and Voltaics for Radioisotope Microbatteries* (Eds. K.E. Bower, et al.), 2002, p. 69, (CRC Press, New York).

[5] Olsen, L.C., Advanced Betavoltaic Power Sources, *Proceeding of the 9th Intersociety Energy Conversion Engineering Conference (IECEC)*, pp. 754 – 762, San Francisco, CA, August 26 – 30, 1974.

[6] U.S. Patent 5124610, Tritiated Light Emitting Polymer Electrical Energy Source, Issued June 23, 1992.

[7] Kavetsky, A.G., et al., Conversion of Radioactive Decay Energy to Electricity. In *Polymers, Phosphors, and Voltaics for Radioisotope Microbatteries* (Eds. K.E. Bower, et al.), 2002, p. 9, (CRC Press, New York).

[8] Bockriss, J. O. and Srinivasan, S., "Fuel Cells: Their Electrochemistry," McGraw-Hill Book Company, pp.607 – 612, 1969.

[9] Eerkens, W. and Reder, M.C., "Closed-Cycle Monopropellant Fuel Cell System Employing Radiation-Generated or Photochemically Generated Ozone," Paper No. 1306-60, ARS Space Power Systems Conference, September 27 – 30, 1960.

[10] Dyer, C.K., A Novel Thin-Film Electrochemical Device for Energy Conversion, *Nature*, 343, pp. 547 – 548, 1990.

[11] Hibino, T., Hashimoto, A., Inoue, T., Tokuno, J-I., Yoshida, S-I., Sano, M., A Low-Operating-Temperature Solid Oxide Fuel Cell in Hydrocarbon-Air Mixtures, *Science*, 288, pp. 2031, 2000.